

DOCKET NO: 271496US0PCT

IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF :
YOSHIKAZU OGURA, ET AL. : EXAMINER: WEIER, A.J.
SERIAL NO: 10/532,727 :
FILED: APRIL 27, 2005 : ART UNIT: 1794
FOR: METHOD OF REMOVING CAFFEINE FROM CAFFEINE-CONTAINING
CATECHIN COMPOUND COMPOSITION

DECLARATION UNDER 37 C.F.R. § 1.132

COMMISSIONER FOR PATENTS
ALEXANDRIA, VIRGINIA 22313

Sir:

Now comes Masahiro Fukuda who deposes and states that:

1. I am a graduate of Tokyo University of Technology and Agriculture and received my Masters degree in the field of Chemistry in the year 1986.
2. I have been employed by Kao Corporation, for 20 years in Research and Development.
3. I understand the English language or, at least, that the contents of the Declaration were made clear to me prior to executing the same.
4. The following experiments were carried out by me or under my direct supervision and control.

5. The following experiment was performed to based on the method disclosed in JP 6-128168. JP 6-128168 and JP 46-39058 disclose a method which entails contacting an alcoholic extract liquor of tea leaves with activated carbon (and acid clay or activated clay). It is questionable whether these methods are sufficient to improve the color tone (or hew) of a beverage, and also whether these methods are capable of efficiently removing caffeine from such compositions. To address these questions, the following experiments were conducted.

6. The experiments in this Declaration were conducted according to the following protocol:

Samples of green tea extracts were prepared according to the method described in Example 1 of JP 6-128168, and were evaluated in some respects, such as hew and the weight ratio of non-polymer catechins/caffeine after treatment, in comparison with the green tea extracts of the present invention.

Green tea leaves (KU-1; 100 g) were immersed in a 99.5% aqueous solution of ethanol (600 g) at 25°C and allowed to undergo an extraction step for 2 minutes. After that, the solution was filtered by a mesh to filter out the tea leaves and further squeezed under pressure. To the ethanol solution thus obtained (560 g) were added 4 g of activated carbon("KURARAY COAL GLC", product of Kuraray Chemical Co., Ltd) and 4 g of acid clay ("MIZUKA ACE #600", product of Mizusawa Industrial Chemicals Ltd.), and this mixture was subjected to a batch treatment for 10 minutes, followed by carrying out a suction filtration through No. 2 filter paper. Subsequently, the mixture was concentrated by a rotatory evaporator and then subjected to water displacement, and the concentration was further continued until the content of the tea extract becomes 35 wt.%. The tea extract was allowed to be freeze-dried over a day and diluted with water (dilution rate: 0.2 g of tea extract per 100 ml of water), and then filtered through a 0.45 μ m membrane filter. The solution thus passed

through the membrane filter was subjected to high-performance liquid chromatography (HPLC) to measure the amounts of both catechin and caffeine. The measurement is conducted in the same manner as described in the present application. The results were shown in the Table* appearing in paragraph 7 below.

(* : The data pertaining to Examples 2 and 3 and Comparative Examples 3 and 4 appearing in this table were reproduced from Table 2 of the present application, while the data of Comparative Example 5' were freshly obtained by this experiment.)

7. The results are shown in the following table:

In the following table, the following correspond to the designated footnotes and is taken from page 43 of the specification for the above-identified application.

1) Composition of non-polymer catechins in “POLYPHENONE HG” preparation:

GC (gallocatechin): 6.39%, EGC (epigallocatechin): 29.42%, C (catechin): 2.16%, EC (epicatechin): 10.3%, EGCg (epigallocatechin gallate): 37.13%, GCg (gallocatechin gallate): 1.93%, ECg (epicatechin gallate): 11.89%, Cg (catechin gallate): 0.79%; content of gallates: 51.73%, content of gallocatechins: 74.88%.

2) Amount of caffeine when “POLYPHENON HG” was dissolved in water: 30.1 mg/100 g. Polyphenon HG is a caffeine-containing composition that is manufactured by Tokyo Food Techno Co., Ltd., and is commercially available from its parent company Mitsui Norin Co., Ltd. Attached to this Declaration is a certificate, with English translation, obtained from Mitsui Norin Co., Ltd. in which it is certified that “POLYPHENON HG” is a tea extract obtained by subjecting tea leaves to a hot water extraction.

Application No. 10/532,727
Declaration under 37 C.F.R. § 1.132

| | Example | Example | Comp. Ex. | Comp. Ex. | Comp. Ex. |
|---|---|---|--|--|--|
| | 2 | 3 | 3 | 4 | 5' |
| Caffeine-containing catechin composition (g) ("POLYPHENON HG", product to Tokyo Food Techno Co., Ltd) | 100 | 300 | 100 | 100 | — |
| Ethanol (g) | 630 | 825.8 | 0 | 0 | — |
| Water (g) | 270 | 374.2 | 900 | 900 | — |
| Activated carbon (g) ("KURARAY COAL GLC", product of Kuraray Chemical K.K.) | 20 | 30 | 0 | 20 | — |
| Acid clay (g) ("MIZUKA ACE #600", product of Mizusawa Chemical Industries, Ltd.) | 100 | 30 | 100 | 100 | — |
| Organic solvent/water (weight ratio) | 70/30 | 71/29 | 0/100 | 0/100 | |
| Amounts of non-polymer catechins after treatment (wt%) ¹⁾ | | | | | |
| GC (gallo catechin) | 6.81 | 7.21 | 6.75 | 7.42 | 4.00 |
| EGC (epigallocatechin) | 32.08 | 29.74 | 31.75 | 34.72 | 38.70 |
| C (catechin) | 2.09 | 1.36 | 0.22 | 2.02 | 1.10 |
| EC (epicatechin) | 8.96 | 9.38 | 9.64 | 8.24 | 10.10 |
| EGCg (epigallocatechin gallate) | 37.12 | 37.76 | 35.93 | 35.86 | 36.50 |
| GCg (gallo catechin gallate) | 1.39 | 1.80 | 1.39 | 1.47 | 0.90 |
| ECg (epicatechin gallate) | 10.78 | 11.31 | 11.34 | 9.61 | 8.50 |
| Cg (catechin gallate) | 0.75 | 0.45 | 0.94 | 0.66 | 0.20 |
| Non-polymer catechins/caffeine after treatment (weight ratio) | 38.8 | 8.8 | 23.6 | 42.6 | 4.55 |
| Amount of caffeine after treatment ²⁾ (mg/100 mL) | 4.8 | 18.1 | 8.2 | 4.3 | 16.70 |
| Content of gallates in non-polymer catechins after treatment (wt%) | 50.0 | 51.3 | 49.6 | 47.6 | 46.10 |
| Content of gallo catechins in non-polymer catechins after treatment (wt%) | 77.4 | 76.5 | 75.8 | 79.5 | 80.10 |
| Absorbance(-) | 0.044 | 0.073 | 0.535 | 0.270 | 0.312 |
| Evaluation of purified product | Caffeine was decreased, and hue and stability were both good. | Caffeine was decreased, and hue and stability were both good. | Hue was deteriorated, and a precipitate occurred | Hue was deteriorated, and a precipitate occurred | Hue was deteriorated, and a precipitate occurred |

8. As evidenced by Comparative Example 5' in the table appearing in paragraph 7 above, the purified product of alcoholic extract of tea leaves shows its absorbance to be 0.312, which is indicating the evidence that the hue of the product is extremely deteriorated, compared with the ones provided by Examples 2 and 3. Furthermore, the weight ratio of non-polymer catechins/caffeine after treatment is 4.55, from which it can also be said that the removal of caffeine is almost impossible where a tea extract is produced according to Comparative Example 5'.

Thus, it is concluded that the process of the present invention is significantly superior to the one described in JP 6-128168, by virtue of its specifically claimed multiple steps, which comprises dissolving a caffeine-containing catechin composition a 9/1 to 1/9 by weight mixed solution of an organic solvent and water, and then bringing the resultant solution into contact with activated carbon and allowing said activated carbon to absorb caffeine to remove caffeine.

9. I declare further that all statements made of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

10. Further Declarant saith not

Masahiro Fukuda
Name: Masahiro Fukuda

August 5, 2009
Date

2009 年 7 月 9 日

花王株式会社 御中



証明書

～ポリフェノン HG の抽出法に関して～

拝啓 貴社益々ご清栄の段お慶び申し上げます。
平素より格別のご愛顧を賜り厚く御礼申し上げます。
さてお問合せ頂きました弊社製品「ポリフェノン HG」の抽出法に関して、以下の通りの工程であることを記します。
ご確認のほど、宜しくお願い申し上げます。

敬具

記

抽出法： 熱水抽出

ポリフェノン HG は、緑茶葉からの熱水抽出物であることを証明いたします。

以上

KAO CORPORATION, Esq.

CERTIFICATE

In re the extraction method of POLYPHENON HG

Dear Sirs,

We are very pleased to hear that you are prospering more and more in business. We would like to express our appreciation for your royal patronage continuously extended to us.

We have heard that you are in need of a certificate pertaining to the extraction method used to obtain "POLYPHENON HG", which is a product of our company. In response to your request, we inform that this extraction method is as follows:

Hot water extraction

We hereby certify that "POLYPHENON HG" is a tea extract which was obtained by applying tea leaves to a hot water extraction method.

Date: July 9, 2009

Mitsui Norin Co., Ltd. (sealed)